



Fig. 1. The Allied-General Nuclear Services spent-fuel reprocessing plant in South Carolina.

Dynamic Materials Accounting Systems

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The problem of maintaining strict control over nuclear material will be made more difficult by the nuclear power demands of the future, which will require large facilities—enrichment plants and reprocessing plants, for example—that process great quantities of high-quality fissile materials. The scale of these operations has forced a reassessment not only of facility design, construction, and process operation, but also of safeguards methods to prevent unauthorized use of the nuclear materials contained in the facilities. A comprehensive domestic safeguards system combines the functions of materials accounting and physical protection.

The Los Alamos Scientific Laboratory has been designated the Department of Energy's lead laboratory for the design and evaluation of materials accounting systems for nuclear facilities of the future. In this article, we examine these systems and the techniques for their design. Nuclear materials accounting systems must keep track of large quantities of materials as they

Systems analysis suggests that near-real-time materials accounting systems designed for future large-throughput nuclear facilities can meet high performance standards.

move through the various processing stages and must keep track of them so well that the absence of even small amounts can be detected. The uncertainties inherent in any measurement process and the difficulties of measuring in high-radiation fields behind heavy shielding complicate this task.

We also illustrate the potential benefits of these systems by describing the development and expected performance of a materials accounting system we have designed for the Allied-General Nuclear Services (AGNS) spent-fuel reprocessing plant at Barnwell, South Carolina (Fig. 1). This plant was designed to process large amounts of irradiated fuel from power reactors. The accounting system was designed after the plant was built and with simulated data because the plant is not yet operating.

The potential of system performance is based on projected measurement capabilities of instruments, some of which are still under development. These projections cannot be tested without access to an operating facility. However, our preliminary evaluations suggest that we can design dynamic materials accounting systems for large bulk-processing facilities that meet detection standards close to those recommended by the IAEA.

The Basis for Materials Accounting

The ultimate aim of nuclear safeguards is to be able to state with confidence, "No significant amount of nuclear material has been diverted." The philosophy underlying the development of materials accounting systems is that the truth of the statement can and should be verified. Thus, materials accounting systems are designed to account for or keep track of the amounts and locations of sensitive nuclear materials by periodic measurements. Materials balances are drawn about suitable areas of the facility according to the equation

$$\begin{aligned}\text{Materials balance} = & \text{initial inventory} \\ & + \text{transfers in} \\ & - \text{transfers out} \\ & - \text{final inventory}\end{aligned}$$

defined over a reasonable time interval. In principle, if all nuclear material in each term of the equation has been measured, the materials balance should be zero in the absence of diversion. In practice, however, it is never zero because of the uncertainties inherent in all measurement processes. The measurement uncertainties produce a corresponding uncertainty in the materials balance, so statistical techni-

ques are used to decide whether a balance indicates diversion of material.

At present, materials balances are drawn around an entire plant or a major portion of it after the facility has been shut down and cleaned out to inventory the material present. Although such accounting methods are essential to safeguards control of nuclear materials, they have inherent limitations in sensitivity and timeliness. The sensitivity is limited by measurement uncertainties that may conceal losses of significant quantities of nuclear material in large plants. The timeliness is limited by the frequency of physical inventories; that is, the practical limits on how often a facility can be shut down for inventory and still remain productive.

Both sensitivity and timeliness can be improved by implementation of *dynamic materials accounting*. This approach combines conventional chemical analysis, weighing, and volume measurements with the on-line measurement capability of NDA (nondestructive assay) instrumentation to provide rapid and accurate assessment of the locations and amounts of nuclear material in a facility. Materials balances are drawn without shutting down the plant: in-process inventories are measured, or otherwise estimated, while the process is operating.

To implement the approach, the facility is partitioned into several discrete accounting areas. Each accounting area contains one or more chemical or physical processes and is chosen on the basis of process logic and the ability to draw a materials balance, rather than on geography, custodianship, or regulatory requirements. By measuring all material flows in each area separately, quantities of material much smaller than the total plant inventory can be controlled on a timely basis and any discrepancies can be localized to the portion of the process contained in the accounting area.

Control by dynamic materials accounting is rigorous. It forces a potential divertor to steal nuclear material in quantities small enough to be masked by measurement uncertainties. Thus, to obtain a significant quantity of material, the divertor must commit many thefts and run the concomitant high risk of detection by the accounting system, surveillance instruments, and physical protection system.

Designing a Materials Accounting System

The performance, or diversion detection sensitivity, of a materials accounting system depends on the details of the measurement system, which in turn depend on the details of the process. Because these details vary from one plant to another, the Los Alamos safeguards systems studies focus on specific designs of existing or planned nuclear facilities.

The first step in the development of a facility's accounting system is to determine the flows of nuclear materials through the facility from design data and operator experience. Then, the facility is partitioned into logical accounting areas, and an appropriate measurement system is postulated for each area. Wherever

possible, the designer incorporates the measurement processes already in the plant design into the measurement system and augments them with any additional measurements necessary to draw a materials balance.* The final step is to examine the expected performance of the accounting system design.

To develop preliminary designs of materials accounting systems, we model and simulate the in-plant processes and measurement systems by computer because no large fuel-cycle plants are yet in operation. Detailed dynamic models of material flows are based on actual process design data. They include bulk flow rates, concentrations of nuclear materials, holdup of materials in the process line, and the variability of all these quantities. Design concepts for the accounting systems are evolved by identifying key measurement points and appropriate measurement techniques, comparing possible materials accounting strategies, developing and testing appropriate data-analysis algorithms, and quantitatively evaluating the proposed system's capability to detect losses. The use of modeling and simulation allows us to study the effects of process and measurement variations over long operating periods and for various operating modes in a short time.

Computer codes simulate the operation of each model process using standard Monte Carlo techniques. Input data include initial values for all process variables and values of statistical parameters that describe each independent process variable. These data are best estimates obtained from process designers and operators. Each accounting area is modeled separately. When a process event occurs in a particular area, the values of the flows and in-process inventories associated with that part of the process are computed

and stored in a data matrix. These data are available for further processing and as input to computer codes that simulate accounting measurements and materials balances.

The flow and inventory quantities from a simulated process model are converted to measured values by applying simulated measurements. Each measurement type is modeled separately; measurement errors are assumed to be normally distributed (Gaussian), and provisions are made for both additive (absolute) and multiplicative (relative) errors. Significant measurement correlations are included explicitly. In most cases the measurement models are derived from the performance of similar instrumentation that has been used and characterized in laboratory and field applications involving similar materials. Simulated measurements are combined to form dynamic materials balances under various accounting strategies.

Data Analysis

We combine the most promising measurement and accounting strategies with statistical techniques in comparative studies of loss-detection sensitivities. One of the major functions of the materials accounting system is to indicate loss, or possible diversion. Diversion may occur in two basic patterns: abrupt diversion (the single theft of a relatively large amount of nuclear material) and protracted diversion (repeated thefts of nuclear material on a scale too small to be detected in a single materials balance because of measurement uncertainties). Protracted diversion usually is the most difficult to detect.

The use of dynamic materials accounting enhances the ability to detect both diversion patterns, but it results in the rapid accumulation of relatively large quantities of materials accounting data. For example, if an area's materials

*See "Nondestructive Assay for Nuclear Safeguards."

balance is closed once each 8-hour shift, after 1 month the safeguards operator will have a sequence of 84 materials balances and estimates of their associated uncertainties. Analysis of a single materials balance may be sufficient to detect a large abrupt theft of material, but the entire sequence of data contains the information necessary to detect small protracted diversions. Because small diversions may be masked by measurement uncertainties, they often are difficult to detect, and the operator must use one or more statistical tests of the accounting data to decide whether diversion has taken place.

Decision Analysis

We have developed or adapted a variety of statistical tools for the analysis of materials accounting data that become available sequentially in time. These tools and their implementation are known collectively as *decision analysis*.

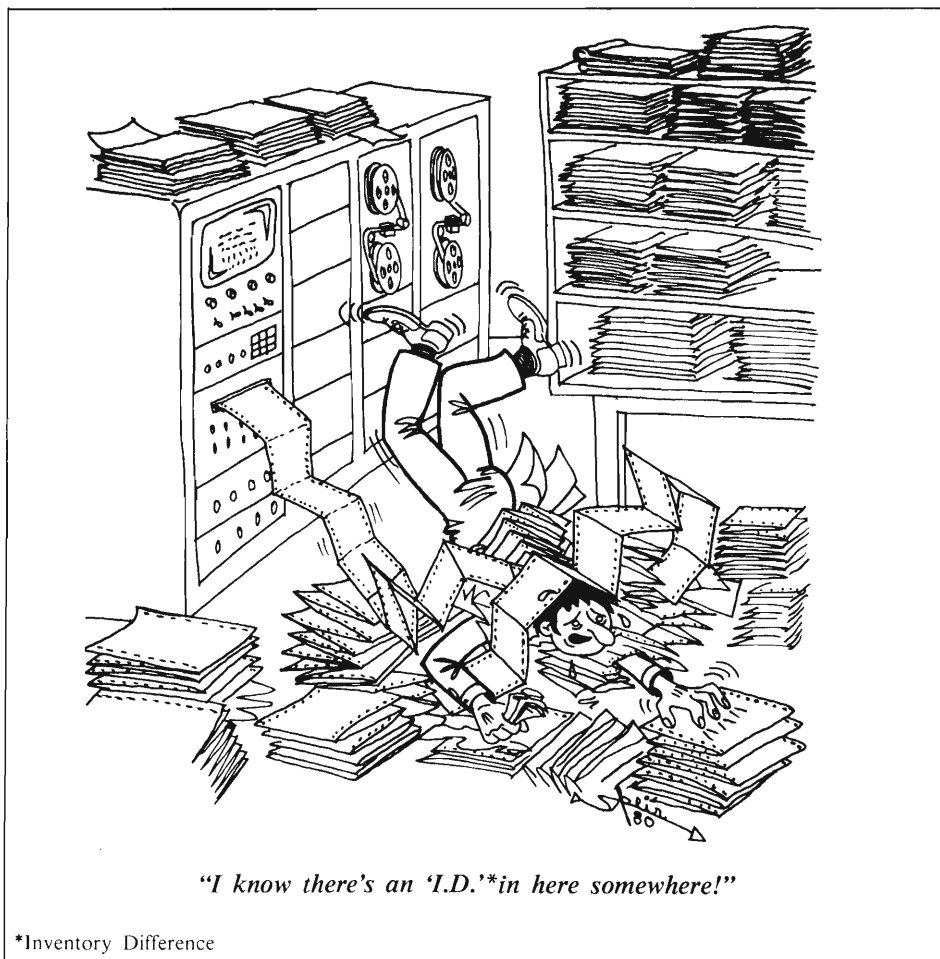
A simple, specific example shows what is involved in decision analysis. Suppose we have a sequence of 10 materials balances (47, 2, -109, 76, 2, 40, 62, -20, 34, 18 g)* and an estimate of each balance's standard deviation. The standard deviation σ is a measure of

the uncertainty in a materials balance calculated from individual measurement errors. To analyze these data we must select an appropriate *statistical test*, construct a *test statistic* from the materials balance data, and establish one or more *test thresholds*. Then we can compare the value of the test statistic to the threshold(s) and draw a conclusion as to whether material has been diverted.

In our example, we use the *cusum test*, a statistical test that uses the cumulative summation (cusum) of the materials balances as the test statistic. The cusum is used often because it provides an estimate of the total amount of material diverted during an accounting period. Other test statistics include a single materials balance or a weighted average of the materials balances. Our cusum test will have a single test threshold determined by the false-alarm probability—that is, the probability of concluding (because of measurement uncertainties) that nuclear material has been diverted when, in fact, no diversion has occurred. The false-alarm probability (FAP) is a measure of the significance of the test results. The FAP value used in setting up the test usually depends on the false-alarm rate that can be tolerated in the plant. The rate often depends on the consequences (shutting down the plant, perhaps) of incorrectly concluding that diversion has taken place.

Our cusum test is illustrated in Fig. 2. In the absence of diversion, we would expect the value of our test statistic—the cusum—to be zero. However, because of measurement uncertainties, the cusum value we get from our accounting data will almost never be zero. The curve in Fig. 2 represents the probability distribution of getting various cusum values when no diversion has occurred. The curve is centered at zero—the expected cusum value. The total area under the curve is 1 because the probability is 100% that the cusum has *some* value.

*The values are the result of a Monte Carlo simulation: they were obtained from a sequence of 10 normally distributed random numbers having mean zero and standard deviation 1 by using the relationship $MB = RN \times \sigma + D$, where MB is the materials balance, RN is a random number, and D is the diversion.



The width of the curve is determined by the uncertainty (measured by the standard deviation σ) in the cusum, which can be computed from the uncertainties in the individual materials balances. In our example, the standard deviation of the cusum is 100 g. The area under the curve to the left of any cusum value represents the probability that—in the absence of diversion—the cusum will have this value or less.

Now we must set our test threshold. We assume that a 5% false-alarm rate is acceptable. In this case, the test threshold (labeled Z in Fig. 2) is set at 165 g. The 5% of the area under the curve lying to the right of the threshold represents the false-alarm probability, labeled FAP.

We are finally ready to test our materials accounting data for evidence of diversion. The materials balances in our example sum to 152 g. Because this cusum value is less than our test threshold, we conclude that there has been no diversion.

Had our cusum value been greater than 165 g, we would have concluded that material had been diverted, but we recognize that there is a significant chance that this would be an incorrect conclusion. If there were no other considerations, the false-alarm probability could be reduced to any arbitrarily small number by increasing the value of the test threshold—but then what would happen to our ability to detect the diversion of a significant amount of material? The relationship between the test threshold and the detection probability is illustrated in Fig. 3.

Suppose 250 g of material have been diverted. Our probability curve is now centered at 250 g because this is the expected cusum value under the hypothesis that this amount of material has been diverted. The width of the curve has not changed because our cusum still has the same associated uncertainty, and the test threshold has not moved. The area un-

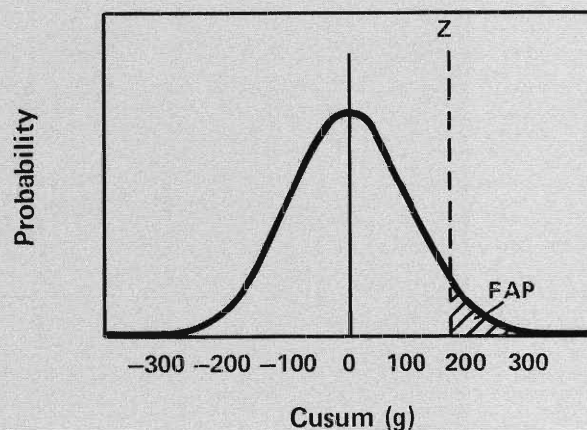


Fig. 2. A cusum test having a single test threshold, $Z = 165$ g. The value of Z is determined by the false-alarm probability (FAP) desired. The FAP is the area under the curve to the right of Z , and in the example, this area is 0.05.

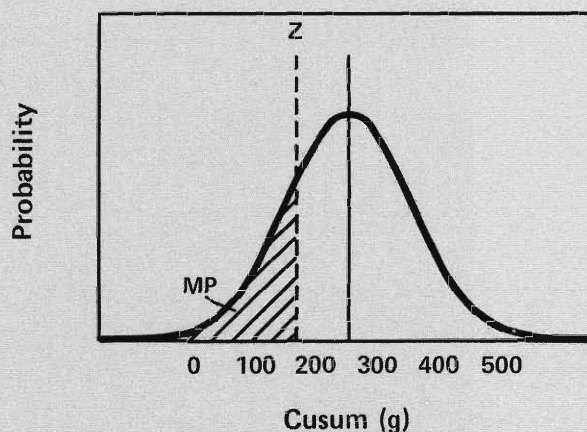


Fig. 3. A cusum test with a total diversion of 250 g and a test threshold kept at $Z = 165$ g. The area under the curve to the right of Z is the detection probability, which, in this illustration, is 0.80. The miss probability (MP) is $1 -$ the detection probability.

der the curve to the right of the threshold represents the probability of detecting the diversion of 250 g of material. (In Fig. 3 this area is 0.8.) The shaded area on the left of the test boundary is the miss probability (MP). It is the probability of concluding that there has been no diversion when, in fact, 250 g have been diverted; it is equal to 1 — the detection probability.

In our example, the cusum is 152 g. This value is smaller than the test boundary, and we have already concluded that there was no evidence of diversion in our materials accounting data. However, if material has been diverted (as illustrated in Fig. 3), we have failed to detect this fact. With our test boundary set for a 5% false-alarm probability, we have only an 80% probability of detecting the diversion of 250 g of material. As the amount of material diverted increases, so does our ability to detect the diversion.

In our example, we considered a single cusum test of 10 materials balances and found that we could choose our test threshold based on an acceptable false-alarm rate. In practice, the 10 balances in our cusum would have been accumulated over a period of time: 10 weeks, if a materials balance were drawn at the end of each week. Perhaps we would like to test each materials balance as it becomes available or test the current cusum as each new materials balance is added. However, if we test each cusum, and if the false-alarm and miss probabilities are fixed for each test, the overall false-alarm and miss probabilities become unacceptably large after several such tests.

Sequential Tests

Another kind of test, the sequential probability ratio test (SPRT) is particularly suited for analyzing data that become available sequentially. The SPRT allows us to guarantee that

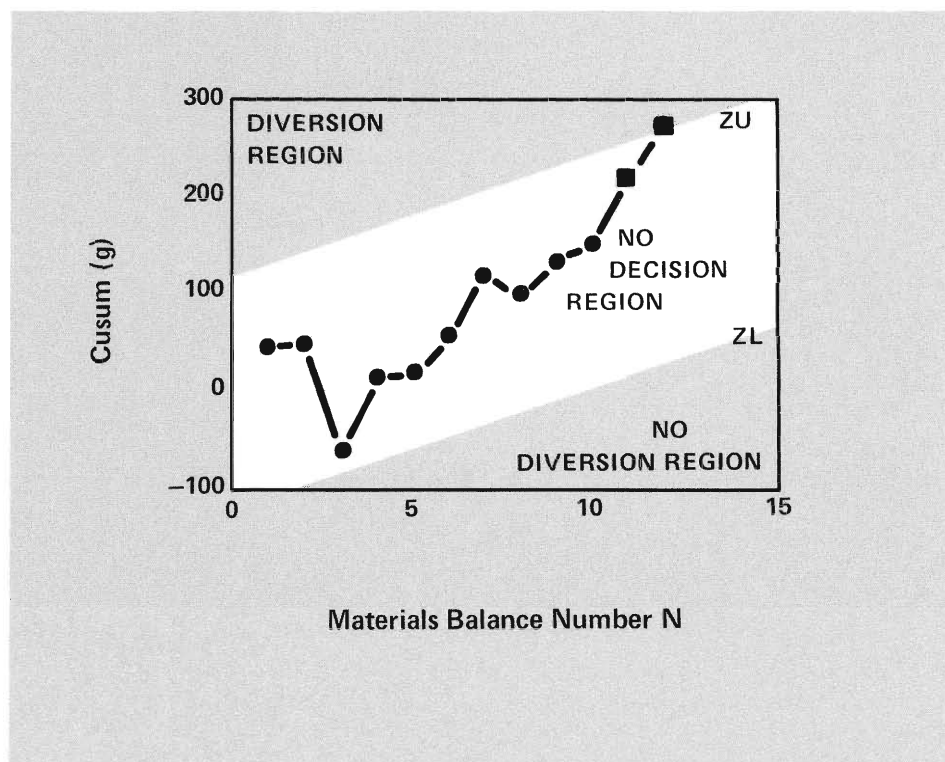


Fig. 4. An example of the Sequential Probability Ratio Test with diversion detected at the 12th materials balance. The test thresholds are established to detect a diversion of 25 g of material during each materials balance period with a detection probability of 95% and a false-alarm probability of 5%. Because the test allows for a no-decision region, an incorrect decision was not made.

neither the false-alarm probability nor the miss probability will exceed desired values, no matter how long the sequence. The cusum remains an appropriate test statistic.

A sequential test has an upper and a lower threshold. Thus, at any time the test result may be that no diversion has occurred, that diversion has occurred, or that no decision can be made until more data are available. Both test thresholds depend not only on the false-alarm probability but also on the desired detection probability, the average rate of diversion, and the number of materials balances in the cusum.

A typical SPRT is illustrated by Fig. 4 for our example sequence of materials balance data. In this test, as each new materials balance is drawn, it is added to the previous cusum to obtain a new cusum value; the value is plotted against the materials balance number. The upper and lower test thresholds are the two parallel lines labeled ZU and ZL, respectively, which divide the cusum chart into three regions indicating diversion, no decision, and no diversion. If the current

cusum value falls above ZU, we conclude that diversion has taken place. If it falls below ZL, there is no evidence of diversion. If it lies between ZU and ZL, we wait for the next materials balance to be drawn. The thresholds have a positive slope because, if a pattern of protracted diversion is present, the total amount of material diverted increases as the number of materials balances in the cusum increases.

The thresholds in Fig. 4 were set for 5% false-alarm probabilities and 5% miss probabilities and for an average 25-g rate of diversion. The settings mean that we would like to detect, with at least 95% probability, the removal of 25 g of material during each balance period and that we can tolerate a false-alarm rate no greater than 5%. The circular symbols correspond to the 10 cusums computed from our example data sequence. The 10th cusum ($N = 10$) lies in the region between the test thresholds so, at the time the 10th materials balance was drawn, we were unable to make a decision. Earlier, we saw that the single cusum test applied after this materials

balance resulted in the conclusion that no diversion had occurred. Indeed, a similar test applied to each of the nine previous cusums would have resulted in the same conclusion. Such conclusions are incorrect for the simulated process from which our example sequence of materials balances was taken: 25 g had been diverted from each materials balance. On the other hand, the SPRT still has not permitted a decision after the 10th materials balance, and thus no incorrect decision has been made. However, after two additional materials balances are drawn, the cusum exceeds the upper test threshold, resulting in the (correct) conclusion that material has been diverted. The current ($N = 12$) cusum value of 271 g provides an estimate of the total amount diverted: the true quantity was 300 g.

Test Statistics

A variety of test statistics can be formed from the materials accounting data and tested sequentially for indications of diversion. Each statistic is based on a different assumption concerning the state of prior knowledge of the measurement errors and of the diversion strategy. Three of the most useful test statistics are the Shewhart, cusum, and uniform diversion statistics.

The Shewhart chart (Fig. 5) is the oldest graphical-display tool to be used widely by industry for process control. In the standard form, measured data are plotted sequentially on a chart where $2\text{-}\sigma$ and $3\text{-}\sigma$ levels are indicated. In safeguards applications, the Shewhart chart is a sequential plot of the materials balance data with $1\text{-}\sigma$ error bars. This chart is most sensitive to large, abrupt shifts in the materials balance data.

The cusum statistic is computed after each materials balance period. It is the sum of all materials balances since the beginning of the accounting interval. Cusum charts are sequentially plotted

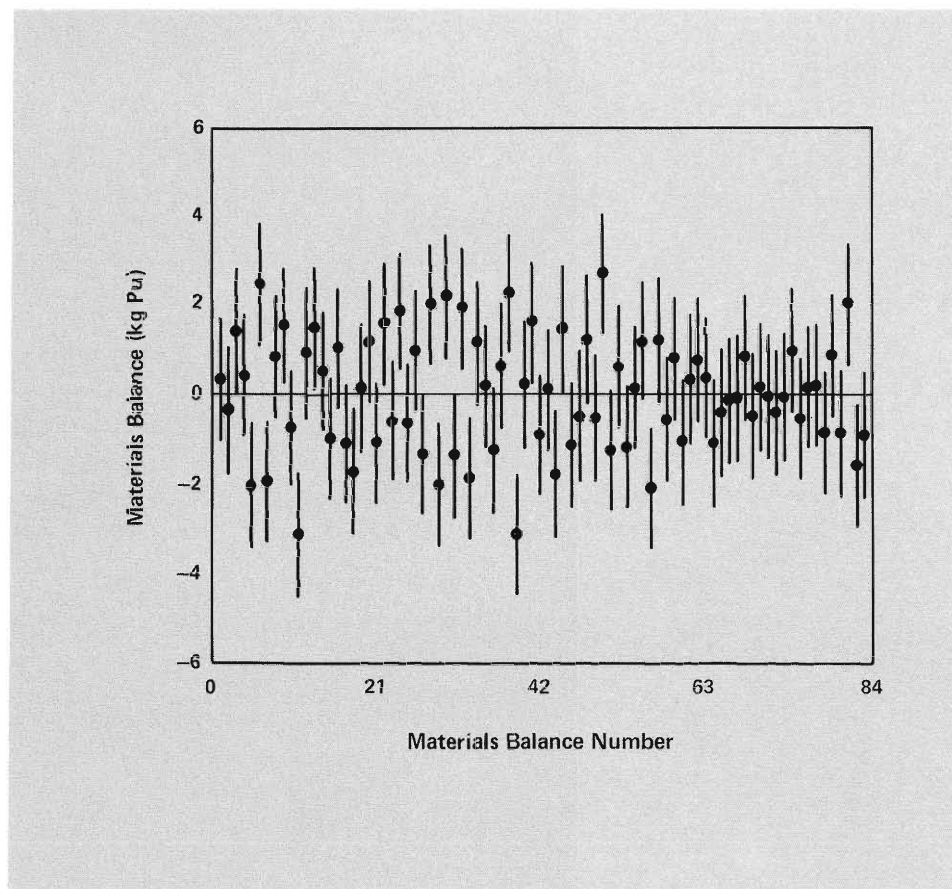


Fig. 5. The Shewhart chart is a graph of sequential materials balance values and their respective materials balance numbers. For each materials balance number, the short, horizontal line gives the materials balance value, and the vertical lines above and below represent the $\pm 1\text{-}\sigma$ deviations from this value. This chart is rather insensitive to protracted, low-level material diversions, but is sensitive to large, abrupt diversions.

cusum values that are used to indicate small shifts in the materials balance data (Fig. 6). The cusum variance (σ_c^2) is a complex combination of the variances of individual materials balances, because these balances usually are not independent. Correlation between materials balances has two principal sources. The first source is the correlation between measurement results obtained by using a common instrument calibration. The magnitudes of the associated covariance terms depend on the magnitude of the calibration error and the frequency of each instrument recalibration; omission

of these terms can cause gross underestimation of the cusum variance. The second source is the occurrence, with opposite signs, of each measured value of in-process inventory in two adjacent materials balances. As a result, only the first and last measurements of in-process inventory appear in the cusum, and only the corresponding variances appear in the cusum variance.

The Kalman filter is a statistical technique applied widely to communications and control systems for signal processing. It is a powerful tool for extracting weak signals embedded in noise.

It has been applied recently to safeguards because dynamic materials accounting systems rapidly generate large quantities of data that may contain weak signals, caused by repeated, small diversions, embedded in the noise produced by measurement errors.

The uniform diversion test (UDT) is designed to detect a small, constant diversion during each materials balance period. Estimates of the average diversion and the inventory at each time are obtained using the Kalman filter. A chart of the UDT is shown in Fig. 7.

The cusum and the UDT are complementary in several respects. The cusum estimates the *total* amount of missing nuclear material at each time step, and its standard deviation is the $1\text{-}\sigma$ error in the estimate of the total. The UDT, on the other hand, estimates the *average* amount of nuclear material missing from each materials balance, and its standard deviation estimate is the $1\text{-}\sigma$ error in the estimate of the average. Thus, both the cusum and the UDT search for a persistent, positive shift of the materials balance data—the cusum by estimating the total and the UDT by estimating the average.

Alarm-Sequence Charts

The decision tests examine all possible sequences of the available materials balance data because, in practice, the time at which a sequence of diversions begins is never known beforehand. Furthermore, to ensure uniform application and interpretation, each test is performed at several levels of significance (false-alarm probability). Thus, it is useful to have a graphic display that indicates the alarm-causing sequences, specifying each by its length, time of occurrence, and significance. One such tool is the alarm-sequence chart, which has proven useful in summarizing the results of the various tests and in identifying trends in the materials accounting data.

Fig. 6. The cusum chart is a graph of the sums of all materials balances drawn from the beginning of an accounting period versus the number of materials balances in the cusum. In this chart, the short, horizontal lines give these cusum values, and the vertical lines represent $\pm 1\text{-}\sigma$ deviations from these values. Because the chart is relatively sensitive to small shifts in materials balance data, it is useful for the detection of protracted, low-level diversion.

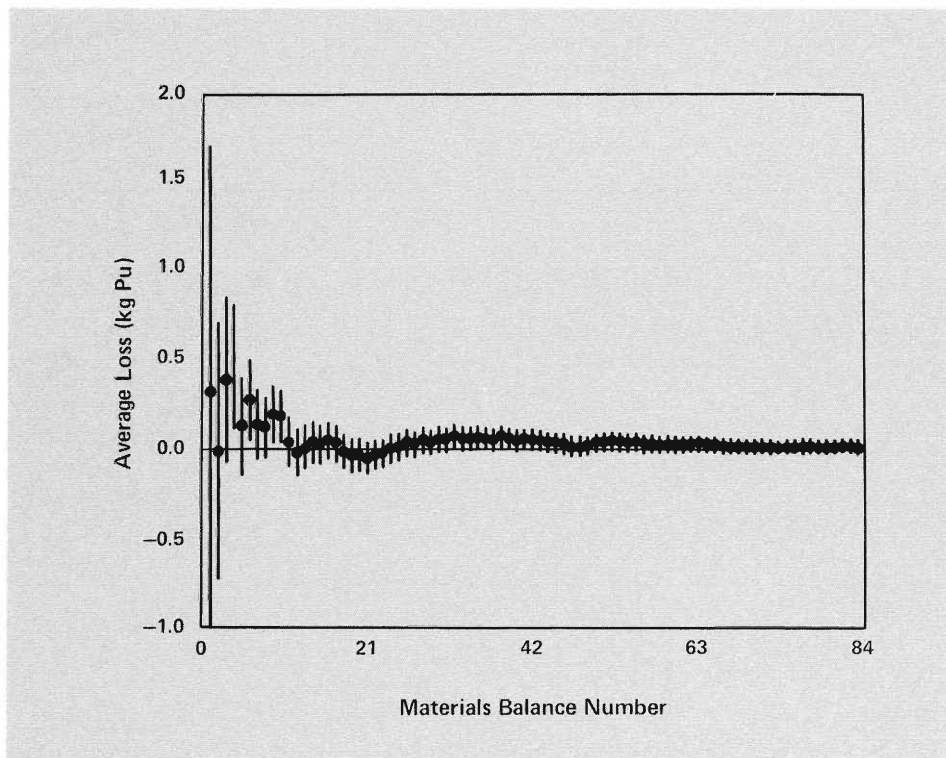
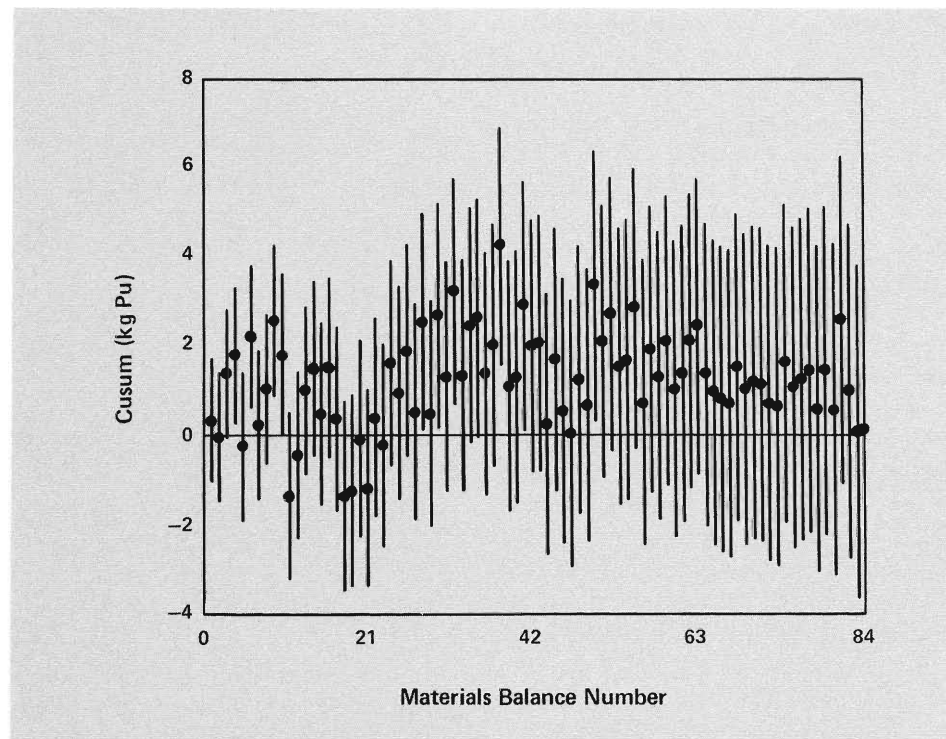


Fig. 7. In this chart of the uniform diversion test, each point on the graph is obtained by a linear combination of previous materials balances. The combination is constructed to provide an estimate of the average amount of material missing per materials balance. Like the cusum test, the UDT searches for a persistent, positive shift in the materials balance data.

An alarm-sequence chart is shown in Fig. 8.

To generate the alarm-sequence chart, each sequence in which the test statistic exceeds the upper boundary ZU and causes an alarm is assigned both a descriptor that classifies the alarm according to its significance (false-alarm probability) and a pair of integers (r_1, r_2) that are, respectively, the indexes of the final and initial materials balances in the sequence. The alarm-sequence chart is a point plot of r_2 vs r_1 for each sequence that caused an alarm, with the significance range of each point indicated by the plotting symbol. One possible correspondence of plotting symbol to significance is given in Table I. The symbol T denotes sequences of such low significance (high false-alarm probability) that it would be fruitless to examine their extensions; the position of the symbol T on the chart indicates the termination point.

TABLE I

Alarm Classification for the Alarm-Sequence Chart

Classification (Plotting Symbol)	False-Alarm Probability
A	10^{-2} to 5×10^{-3}
B	5×10^{-3} to 10^{-3}
C	10^{-3} to 5×10^{-4}
D	5×10^{-4} to 10^{-4}
E	10^{-4} to 10^{-5}
F	$\leq 10^{-5}$
T	> 0.5

It is always true that $r_1 \gg r_2$, so that all symbols lie to the right of the line $r_2 = r_1$ through the origin. Persistent data trends (repeated diversions) cause long alarm sequences ($r_1 \gg r_2$), and the associated symbols on the alarm chart extend far to the right of the line $r_2 = r_1$.

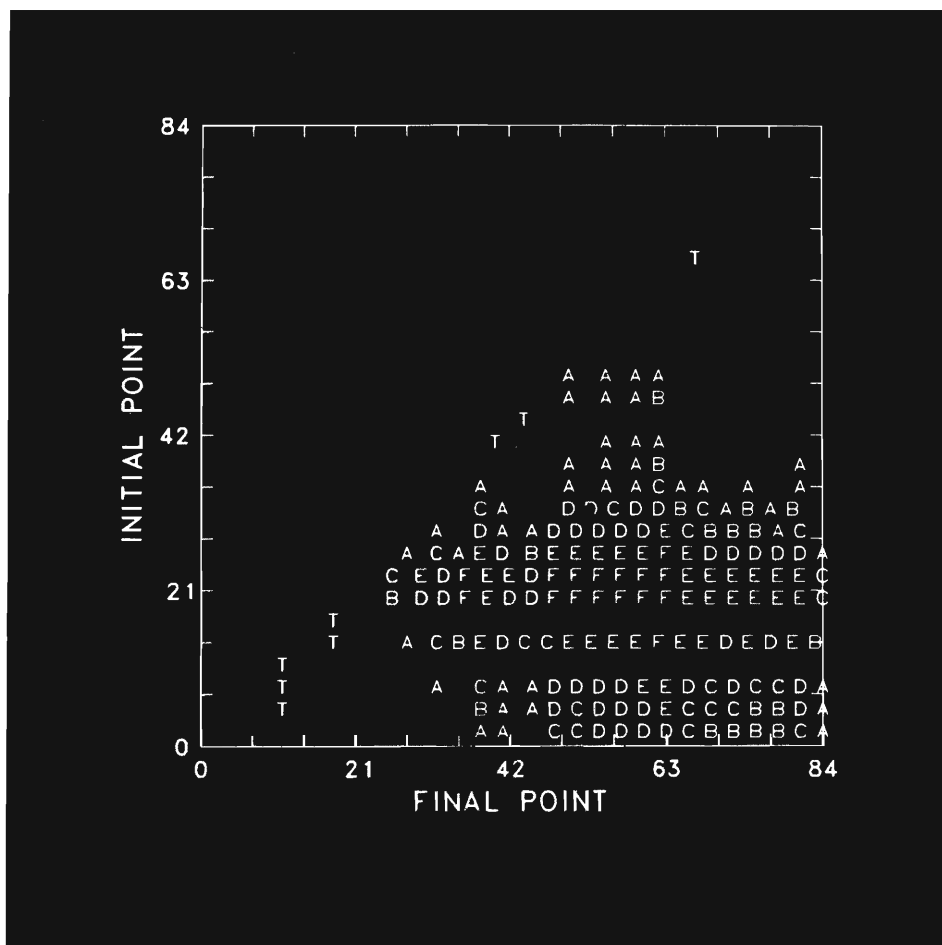


Fig. 8. An alarm-sequence chart. The false-alarm probability associated with each letter is given in Table I. To illustrate how this chart is used, consider a sequence of materials balance data beginning at balance number 21, and suppose that one of the tests gives an alarm with a false-alarm probability of 2×10^{-4} at balance number 30. On the alarm-sequence chart for that test, the letter D would appear at the point (30,21). Because this false-alarm probability is so small, the probability of material diversion commencing with balance number 30 is large.

Systems Performance Evaluation

An analysis of a system's performance in detecting losses of nuclear material is essential to the design of nuclear materials accounting systems. Performance measures must include the concepts of loss-detection sensitivity and loss-detection time. Because materials accounting is statistical, loss-detection sensitivity is described in terms of the probability of detecting some amount of loss while accepting the probability of some false alarms. Loss-detection time is the time required by the accounting system to reach a specified level of loss-detection sensitivity. The loss scenario is not specified in performance measures; whether the loss is abrupt or protracted, the total loss is the measure of performance. The loss-detection time refers

only to the accounting system's internal response time.

The performance of any accounting system can be described by a function

$$P [L, N, \alpha],$$

where P is the accounting system's probability of loss detection, L is the total loss over a period of N balances, and α is the false-alarm probability. A convenient way of displaying system performance is a three-dimensional graph of the surface P vs L and N for a specified value of α , called a *performance surface*. A single point (N, L, P) of such a surface is plotted in Fig. 9. The entire surface portrays the expected performance of an accounting system as a function of the three performance measures, loss, time, and detection probability.

Because systems performance may depend on the details of a particular diversion strategy and, therefore, on the statistical techniques used, overall performance is difficult to quantify. Fortunately, the cusum statistic does not depend on how the material was lost, but responds only to the total loss L during any time interval N . Moreover, even though the cusum test is seldom the best test for any particular scenario, it detects any loss relatively well. Consequently, it is always among the tests applied to the accounting data, and it provides a conservative, scenario-independent measure of systems performance. The performance of more powerful tests for specific loss scenarios, such as the UDT, should be compared with the cusum test performance to ensure that the cusum approximation does not generate undue pessimism.

Measurement Error Models

Because detection sensitivity is limited by measurement errors, we must have measurement models and error estimates for various types of instrumentation to evaluate the performance of a materials accounting system. The simple measurement model given below applies when error standard deviations are expressed on a relative basis and is appropriate for measurement situations in which the associated error tends to be proportional to the quantity being measured.

$$m = M(1 + \varepsilon + \eta), \quad (1)$$

where m is the measured value of a true quantity M .

The measurement errors have been grouped in two categories, instrument imprecision ε and calibration η ; both are regarded as observations on random variables. The instrument imprecision ε represents the deviation of the measured value from the true quantity caused by the scatter or dispersion in a set of in-

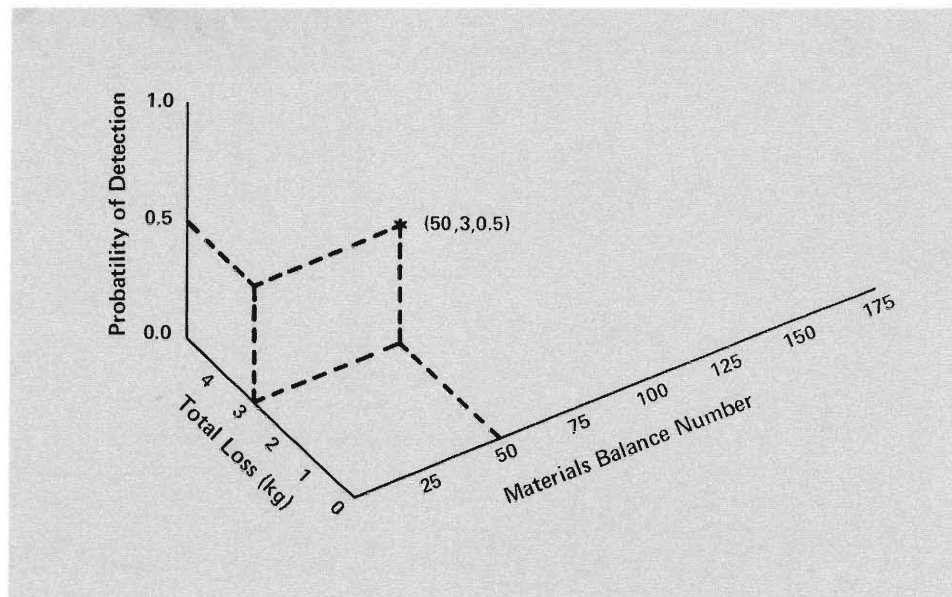


Fig. 9. The three-dimensional space of performance surfaces. Note that the Total Loss axis increases up and to the left. This graph indicates that the probability of detecting a loss of 3 kg of material at balance number 50 is 0.5.

dividual measurement results (for example, the uncertainty caused by counting statistics in NDA measurements). The calibration error η represents the errors that persist, unchanged, throughout a limited set of measurements as a result of the uncertainty in converting raw measurement results into the quantity of interest (for example, in converting counts to plutonium mass for NDA measurements). Calibration errors are the more difficult to estimate because they include uncertainties in standards, calibration parameters, instrument environment, and measurement control procedures.

The error random variables (ε and η) have means of zero and variances of σ_ε^2 and σ_η^2 , respectively. The variance σ_m^2 of the measured value m is given by

$$\sigma_m^2 = M^2(\sigma_\varepsilon^2 + \sigma_\eta^2). \quad (2)$$

To simulate a series of measurements from a given instrument or measurement process, a new value of ε is sampled from the appropriate ε -error distribution for each measurement, whereas a new value of η is sampled from the appropriate η -error distribution only when the instrument is recalibrated. All measurements from the same instrument having the same η error (calibration) are correlated. These correlations become important if an instrument cannot be

recalibrated frequently, and they may dominate the materials balance error. The covariance (a measure of the correlation) between the i^{th} and j^{th} measurements is given by

$$\sigma_{ij} = M_i M_j \sigma_\eta^2. \quad (3)$$

An Ideal Process

A simple example illustrates dynamic materials accounting concepts and principles. Figure 10 represents an ideal process having a daily throughput of 50 kg of nuclear material consisting of twenty-five 2-kg batches. The in-process inventory is 25 kg, and the residual holdup is 5 kg after shutdown and cleanout, which occur once each month. The entire process is contained in a single materials balance area (Fig. 10a); the storage areas for feed and product are located in separate accounting areas (not shown).

Figures 10b and 10c show two possible divisions of the process into accounting areas for dynamic accounting purposes. In Fig. 10b, the process is divided into a *series* of five smaller accounting areas. To use this division, we would measure transfers of nuclear materials between adjacent accounting areas and the in-process inventory in each area. In Fig. 10c, the process is divided into five *parallel* areas

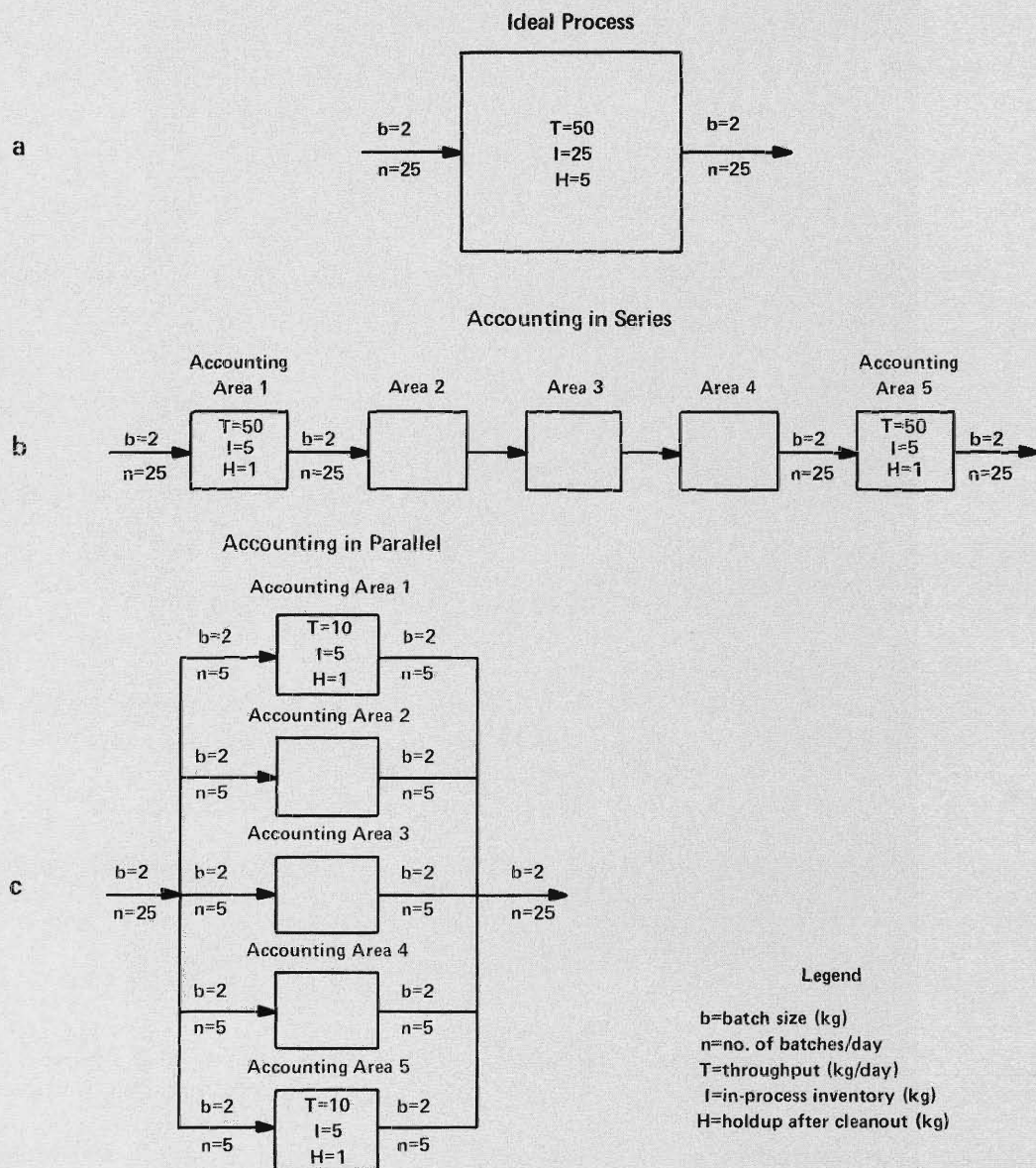


Fig. 10. Materials balance accounting areas for an ideal process. In a, the entire process is contained in a single materials balance area. In b the process is divided into five accounting areas in series; one measurement suffices to determine the transfer out of one area and into the next. The entire daily throughput passes through each area. In c, the five accounting areas are in parallel, that is, the throughput per day in each area is one-fifth of the total throughput; the transfers into and out of each area are measured independently.

corresponding to five separate process lines in parallel. In this case, we would measure the input, output, and inventory of each area separately. In practice, the division of the process depends on its configuration.

We can calculate measurement errors in dynamic materials balances applied to the ideal process by using the measurement model described in Eqs. (1)-(3). For an accounting period during which N batches are processed, the dynamic

materials balance MB_N for one accounting area is given by

$$MB_N = \Delta I_N + T_N, \quad (4)$$

where ΔI_N is the net change in the inventory and T_N is the net transfer of nuclear material (inputs minus outputs) across the accounting area. If there were no measurement errors, MB_N would be exactly zero and, if the process were operated at steady state, ΔI_N and T_N

also would be zero.

Measurement errors produce an uncertainty in MB_N having a variance σ_{MB}^2 (assuming no correlation between transfer and inventory measurements) given by

$$\sigma_{MB}^2 = \sigma_{\Delta I}^2 + \sigma_T^2. \quad (5)$$

Understanding the behavior of the inventory-change and net-transfer variances, $\sigma_{\Delta I}^2$ and σ_T^2 , is basic to the ef-

fective design of a materials measurement and accounting system.

If the initial and final inventories, I_0 and I_N , are measured during the same calibration period (that is, if they have the same η error), the variance $\sigma_{\Delta I}^2$ of the net inventory change ΔI is given by

$$\sigma_{\Delta I}^2 = (I_0^2 + I_N^2) \sigma_{\epsilon I}^2 + (I_0 - I_N)^2 \sigma_{\eta I}^2, \quad (6)$$

where $\sigma_{\epsilon I}^2$ and $\sigma_{\eta I}^2$ are the ϵ - and η -error variances of the inventory measurements. Note that if I_0 and I_N are equal, $\sigma_{\Delta I}^2$ has the minimum value

$$\sigma_{\Delta I}^2 = 2I_0^2 \sigma_{\epsilon I}^2. \quad (7)$$

For a large class of process equipment, efficiency and economy dictate that the in-process inventory be held nearly constant during normal operation. Near-steady-state operation benefits materials accounting by reducing the materials balance uncertainty because the condition $I_0 \cong I_N$ implies that the dependence of σ_{MB} on $\sigma_{\eta I}$ is weak [Eq. (6)]. Hence, a well-known value for $\sigma_{\eta I}$ is not required. This result is important because calibration of in-process inventory measurements may be difficult, especially for process equipment located in high-radiation fields behind heavy shielding. The ideal process is assumed to satisfy the steady-state condition so that Eq. (7) holds. The inventory measurement error ($\sigma_{\epsilon I} = 10\%$ in this example) limits the dynamic accounting sensitivity over short accounting periods.

The variance σ_T^2 of the net material transfer T is given by

$$\sigma_T^2 = 2Nb^2 (\sigma_{\epsilon b}^2 + \sigma_{\eta b}^2) + 2N(N-1)b^2 \sigma_{\eta b}^2, \quad (8)$$

where b is the input and output batch size, and $\sigma_{\epsilon b}^2$ and $\sigma_{\eta b}^2$ are the ϵ - and η -error variances of the batch transfer measurements. For simplicity of presentation,

the error variances of input and output batch measurements have been set equal in value (hence the factor of 2), but the two measurements are statistically independent; that is, they are not correlated.

The first term in Eq. (8) occurs whenever N input and N output batches are measured during the accounting period and is present even if the transfer measurements are uncorrelated. The second term accounts for pair-wise correlations among the transfer measurements [Eq. (3)]. The transfer measurements are correlated primarily because the instruments are not recalibrated during the accounting period. Note that the number of pair-wise correlations increases approximately as N^2 ; if N is sufficiently large, correlations make the dominant contribution to σ_T^2 .

The effect of measurement correlations can be reduced by recalibrating the transfer-measuring instruments. If the instruments are calibrated K times during the accounting period, and if n_k is the number of batches processed between the k^{th} and $(k+1)^{\text{th}}$ calibrations, then σ_T^2 is given by

$$\sigma_T^2 = 2Nb^2 (\sigma_{\epsilon b}^2 + \sigma_{\eta b}^2) + 2b^2 \sigma_{\eta b}^2 \sum_{k=1}^K n_k (n_k - 1), \quad (9)$$

where

$$N = \sum_{k=1}^K n_k. \quad (10)$$

The number of correlation terms in this case increases approximately as $\sum n_k^2$ rather than as N^2 .

The effect on σ_T of daily versus monthly recalibration of transfer-measuring instruments is shown in Fig. 11. The relative standard deviation (RSD), which is σ_T divided by the throughput Nb , is plotted as a function of the number N of processed batches. Values of $\sigma_{\epsilon b}$ and $\sigma_{\eta b}$ have been taken to

be 2% and 0.5%, respectively. The net-transfer RSD varies as $|(\sigma_{\epsilon b}^2 + \sigma_{\eta b}^2)/N|^{1/2}$ for a small N and as $(\sigma_{\eta b}^2/K)^{1/2}$ for a large N , that is, when the transfer correlations are dominant.

Correlations between transfer measurements limit the sensitivity of dynamic materials balances over relatively long accounting periods. Therefore, the parameters $\sigma_{\eta b}$ and K are especially important. The value of $\sigma_{\eta b}$ depends primarily on the measurement control procedures and on the quality of available calibration standards, whereas the value of K depends on how often the transfer-measuring instruments are recalibrated. Adequate measurement controls must include well-characterized standards for the transfer measurements. Further, provision must be made for sufficiently frequent recalibration of the transfer-measuring instruments.

Table II contains kilogram values of the standard deviation σ_{MB} of dynamic materials balances calculated for the ideal process. Results are given for four accounting periods: one batch, 1 day, 1 week, and 1 month (30 days), and for two transfer calibration periods, 1 day and 1 month. The inventory-change and net-transfer components of σ_{MB} are given separately. Calculated values are shown for one accounting area in a

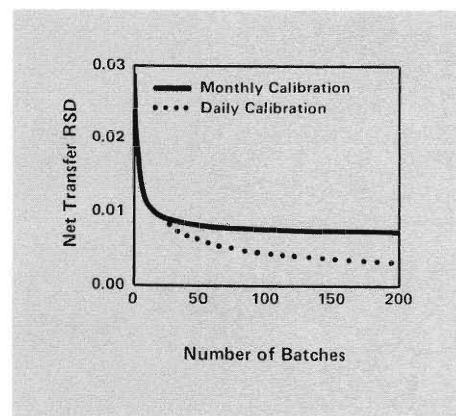


Fig. 11. Effect of calibration on transfer measurement errors.

TABLE II

Dynamic Materials Accounting in an Ideal Process

Accounting Period	Standard Deviation (kg)					
	Monthly Recalibration			Daily Recalibration		
	Series	Parallel	Total	Series	Parallel	Total
1 Batch						
Inventory change	0.71	0.71	1.58	0.71	0.71	1.58
Net transfer	0.06	0.06	0.06	0.06	0.06	0.06
Materials balance	0.71	0.71	1.58	0.71	0.71	1.58
1 Day						
Inventory change	0.71	0.71	1.58	0.71	0.71	1.58
Net transfer	0.45	0.14	0.45	0.45	0.14	0.45
Materials balance	0.84	0.72	1.64	0.84	0.72	1.64
1 Week						
Inventory change	0.71	0.71	1.58	0.71	0.71	1.58
Net transfer	2.59	0.60	2.59	1.20	0.38	1.20
Materials balance	2.68	0.93	3.03	1.39	0.80	1.98
1 Month						
Inventory change	0.14	0.14	0.32	0.14	0.14	0.32
Net transfer	10.72	2.23	10.72	2.48	0.79	2.48
Materials balance	10.72	2.24	10.72	2.48	0.81	2.50

series arrangement, one accounting area in a parallel arrangement, and the total process (see Fig. 10). Note that the data for the total process are a synthesis of the data from the smaller accounting areas. In practical application the capability of combining the same dynamic accounting data in different ways to form materials balances for various accounting envelopes provides obvious safeguards advantages that can be exploited by the materials accounting system software.

The data in Table II support the following conclusions. For relatively short accounting periods, the materials balance standard deviation (σ_{MB}) is determined primarily by the size of the inventory (I) and the inventory instrument-precision RSD (σ_{ei}). For longer accounting periods, σ_{MB} is determined by the sizes of the transfers (b), the transfer calibration-error RSD (σ_{nb}), and the number (K) of transfer-instrument recalibrations.

Reduction of in-process inventory and accessibility of process equipment for inventory measurements are important design considerations. Since the use of parallel process lines reduces throughput and inventory in each accounting area for the same total plant throughput, it often can markedly improve materials accounting sensitivity. This practice, however, requires independent instrumentation for each accounting area. Large-capacity tanks present special accounting problems, and strict surveillance (process monitoring) measures, in addition to materials accounting measures, should be considered for them. Processing relatively small batches and operating the process near steady state generally enhance the capability of materials accounting.

Materials measurements require rapid in-line or at-line assay techniques that provide precise inventory measurements and accurate transfer measurements, and provision for frequent recalibration

of the transfer-measuring instruments. The period between physical inventories should be coupled to the buildup of transfer-measurement correlations; that is, after the materials-balance error standard deviation for the accounting area becomes unacceptably large, a physical inventory is necessary to "restart" the dynamic accounting system.

Application to a Reprocessing Plant

We have applied the principles of a dynamic materials accounting system to a real plant, the fuel reprocessing facility built by Allied-General Nuclear Services (AGNS) at Barnwell, South Carolina. Since this plant is not yet operating, process and materials balance data are simulated for analysis. AGNS is designed to receive and process irradiated power-reactor fuel containing ^{235}U and plutonium. The plant capacity, which is 50 kg plutonium/day, is typical of plants that will be required in the 1990s to support a mature nuclear industry. The AGNS plant uses the Purex recovery process, a process in large-scale use for over 25 years and still used, with minor variations, by most of the reprocessing plants now operating or planned throughout the world. The products are concentrated uranyl nitrate and plutonium nitrate solutions.

Spent-fuel assemblies arrive at the plant by rail or truck and remain in a fuel-storage pool while awaiting processing. The fuel elements are chopped into small pieces, and the fuel is dissolved with a concentrated nitric acid solution. Following dissolution, a paraffin hydrocarbon solvent is used to separate most of the fission products from the plutonium and uranium. The solvent stream containing the plutonium and uranium then enters a partitioning step, where the bulk of the uranium is separated from the plutonium. The uranium stream is further decontaminated, concentrated, and passed

through silica-gel beds to remove traces of zirconium and niobium. The plutonium stream is also further purified, concentrated, and stored to await conversion to plutonium oxide. The wastes from the processes are treated in either liquid- or solid-waste processing systems. Off-gases are treated before being vented to the atmosphere.

Nuclear materials in a fuel-reprocessing facility are present in several different physical and chemical forms and also at different levels of radioactivity. Therefore, the accessibility and desirability of nuclear material for diversion will vary throughout the plant.

We can illustrate this point by the following example. Say we wish to determine the amount and form of material required to divert 1 kg of plutonium from various parts of the process. In the chop and leach portion of the plant, where the fuel enters the recovery process through dissolution in nitric acid, a divertor would need about 330 L of solution to obtain 1 kg of plutonium. Furthermore, because of the fission product content of this solution, the divertor would receive an immediate lethal dose of radiation without the use of very heavy shielding. This portion of the process, then, would be a poor choice for diversion of nuclear material.

If we proceed farther along the process to the chemical separations portion, we find the diversion of plutonium somewhat more attractive. Here the uranium and plutonium are separated from each other, and the fission products are partially removed from solution. The radiation level of the solution in this area is still high, but not immediately lethal. To obtain 1 kg of plutonium, about 200 L of solution must be drained from the storage and sampling tank. This amount is less than that required from the chop and leach portion of the process, but it is still considerable.

Still farther along in the process stream, after chemical concentration of

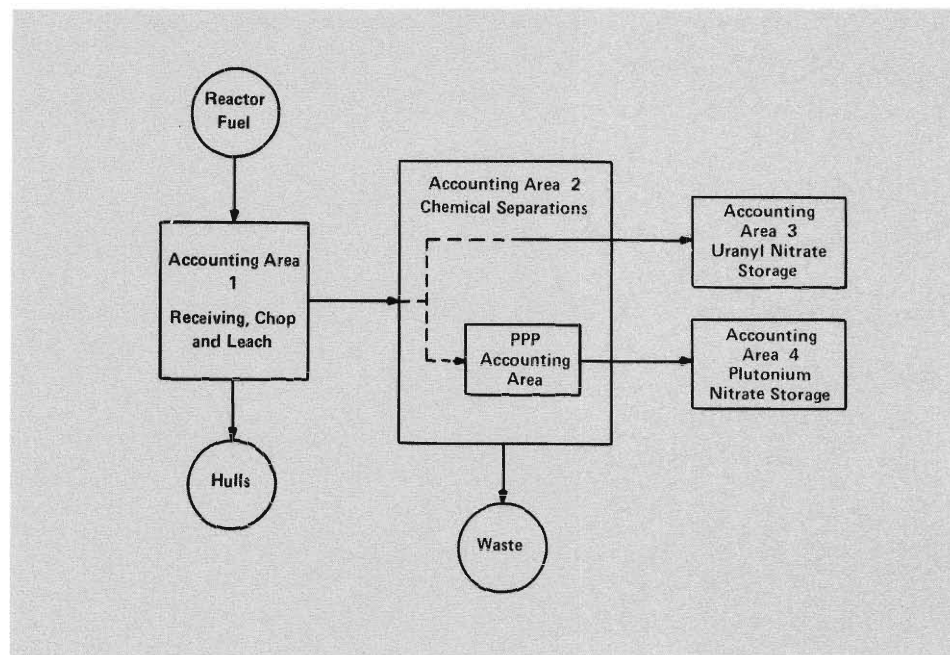


Fig. 12. Accounting areas in the AGNS facility. The plutonium purification process (PPP) accounting area has a total plutonium inventory of about 40 kg and a plutonium throughput of 50 kg/day.

the plutonium nitrate product, only about 4 L of solution would be required to obtain 1 kg of plutonium, and the radioactivity level is so low that no special shielding precautions would be necessary. This portion of the process is especially attractive to a divertor.

The example shows that a *graded* materials accounting system is both useful and economical in developing safeguards for a reprocessing facility. Where the accessibility and attractiveness of nuclear material are low, a safeguards system need not be so stringent. However, the plutonium product near the end of the recovery process is of paramount importance, and rigorous materials control and accounting must be maintained in this area.

Dividing the reference process into several materials accountability areas should be advantageous for materials accounting. For the AGNS facility, we have separated the process into the four accounting areas shown in Fig. 12. In Area 1, fuel is received for storage in pools and, as demanded by the process flow, is removed for chopping and dissolution. The concentrations of nuclear material at the downstream end of this area are about 300 g uranium/L and 3 g plutonium/L. Material of this concentration is transferred to Area 2, where the

plutonium and uranium nitrates are separated, and fission products are removed from solution. At the downstream end of Area 2, product batches of uranium are concentrated to 375 g uranium/L and transferred to Area 3 for storage, while product batches of plutonium are concentrated to 250 g plutonium/L and transferred for storage into Area 4.

For the purposes of this discussion, we will concentrate on the chemical separation process in Area 2 and will restrict our attention to the plutonium purification process (PPP) within that area (Fig. 13 and Tables III and IV).

Materials Accounting for Plutonium Purification Process

Flow and concentration measurements at the 1BP tank (input) and 3P concentrator (output) isolate the PPP as an accounting area. In addition, acid recycles (2AW, 3AW, 3PD) and organic recycle (2BW, 3BW) must be monitored for flow and concentration, and the total in-process inventory must be estimated. Table V gives the required measurement points and some possible measurement methods and associated uncertainties.

The relative precision of dynamic volume measurements is estimated to be 3% (1 σ) for the 1BP tank, threefold

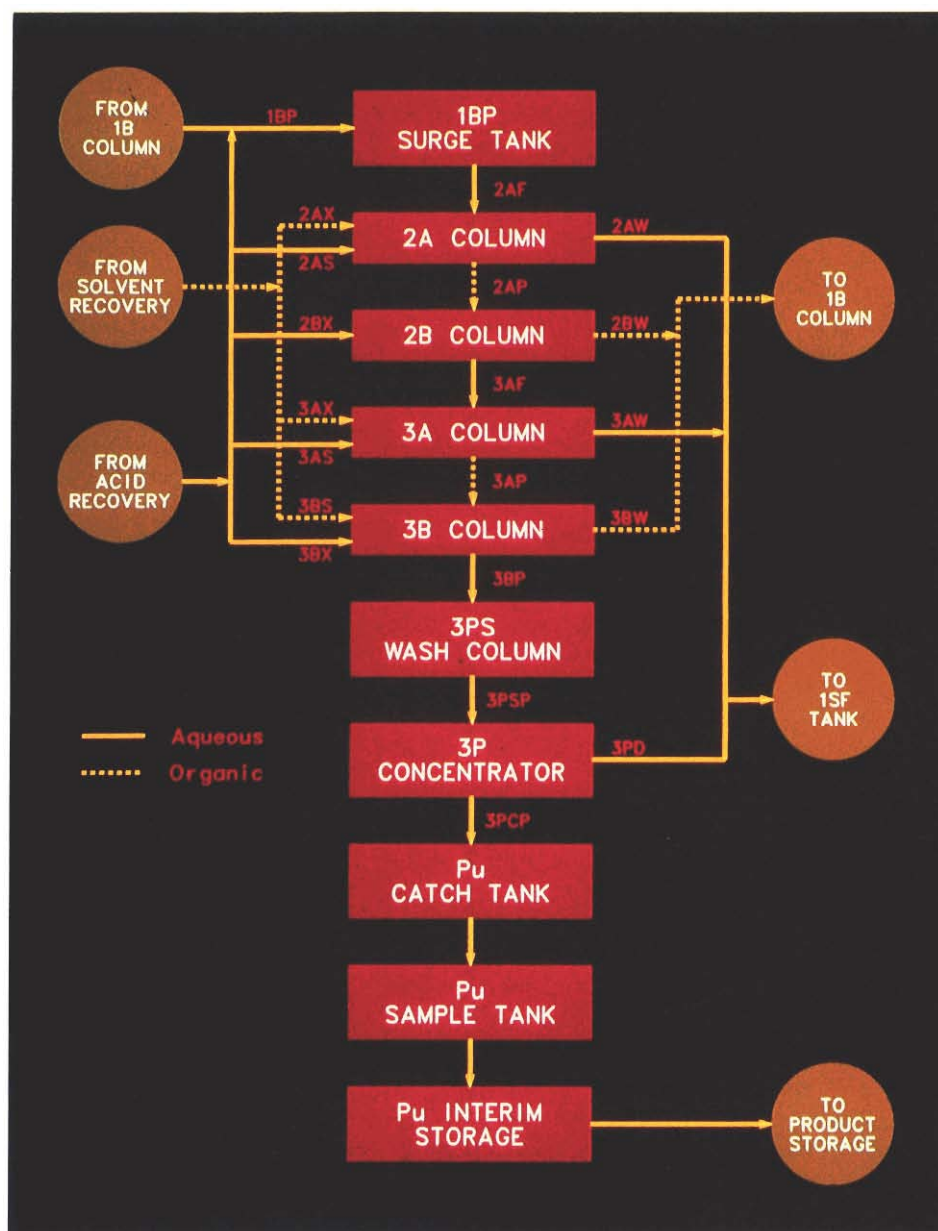


Fig. 13. The plutonium purification process (PPP) accounting area. Tables III, IV, and V describe the plutonium concentrations and flow rates, the in-process inventories, and the materials accounting measurements, respectively, that were used to design and evaluate the performance of a materials accounting system for this area.

more than for a conventional physical-inventory measurement because liquid flows into and out of the tank continuously during processing. Dynamic estimates of plutonium concentration in the 1BP and 3P concentrator tanks can be obtained from direct, in-line measurements (by absorption-edge densitometry, for example) or from combinations of adjacent accountability and process-control measurements.

Pulsed columns 2A and B and 3A and B are used to purify the plutonium. In the AGNS design, the columns are fully instrumented for process control, so that

measurements of plutonium concentration and inventory are possible. Relative precision for column-inventory measurements is estimated to be in the range of 5-20% (1 σ). The 20% limit appears to be conservative in terms of discussion with industry and DOE personnel. A precision of 10% should be practicable with the use of current process-control instrumentation. Improvements toward the 5% figure (or better) will require additional research and development to identify optimum combinations of additional on-line instrumentation and improved models of column behavior.

TABLE III
Concentrations and Flow Rates
in the PPP

Stream	Flow (L/h)	Plutonium Concentration (g/L)
1BP	400	5
3PCP	8	250
2AW	500	trace
3AW	215	0.1
3PD	32	trace
2BW	150	trace
3BW	105	trace

TABLE IV
In-Process Inventories in Tanks
and Vessels of the PPP^a

Identification ^b	Volume (L)	Plutonium Inventory (kg)
1BP tank	1500	7.4
2A column	700	4.6
2B column	500	2.8
3A column	600	5.4
3B column	440	4.8
3PS wash column	20	1.2
3P concentrator	60	15

^aThese values are not flow sheet values of any existing reprocessing facility but represent typical values within reasonable ranges of a workable flow sheet.

^bSee Fig. 13.

Waste and recycle streams from the columns and the concentrator in the PPP are monitored by a combination of flow meters and NDA alpha detectors for plutonium concentration. The alpha monitors are already used for process control in the AGNS design and require only modest upgrading (primarily calibration and sensitivity studies) to be

used for accountability. Flow measurements in the waste and recycle streams can be simple and relatively crude (5-10%) because the amount of plutonium is small.

Because the PPP processes nuclear material semicontinuously, materials balances could be drawn as often as once per hour. However, our studies have shown that an 8-hour balance period gives a reasonable diversion detection sensitivity and matches normal process operating conditions. The following results are based on drawing materials balances every 8 hours.

Performance Evaluation

Simulated results of diversion detection for 1 month of process operation in the PPP accounting area are given in Figs. 14-16. The figures show results obtained with the Shewhart, cusum, and UDT decision analysis tests. Each figure also shows plots of the test statistic and the corresponding alarm chart for the case of no diversion (upper) and for the case of diversion (lower). In the plots of the test statistics, the horizontal marks indicate the values of the statistics, and the vertical lines are 1- σ error bars about those estimates. In each case a strategy of low-level uniform diversion is simulated during the 21st to 63rd materials balances. The Shewhart test is so insensitive to this pattern of diversion that no alarms appear on its alarm chart, while alarms appear on the charts almost immediately after diversion begins for both the cusum and UDT tests.

In the course of evaluation, many such sets of charts are examined so that the random effects of measurement errors and normal process variability can be assessed; that is, we perform a Monte Carlo study to estimate the sensitivity to diversion. However, in applying decision analysis to data from a facility operating under actual condi-

TABLE V

Materials Accounting Measurements for the PPP

Measurement Point	Measurement Type	Instrument Precision (1 σ , %)	Calibration Error (1 σ , %)
1BP, 3PCP streams	Flow meter	1	0.5
	Absorption-edge densitometry	1	0.3
1BP surge tank	Volume	3	a
	Absorption-edge densitometry	3	a
2A, 2B, 3A, 3B columns	See text	5-20	a
2AW, 2BW, 3AW, 3BW, 3PD streams	Flow meter	5	1
3PS column	See text	5-20	a
3P concentrator	Volume (constant)	a	a
	Absorption-edge densitometer	1.5	a

^aNot important.

TABLE VI

Plutonium Purification Process Diversion Detection Sensitivity^a

Accounting Period	Number of Materials Balances	Total at Detection (kg Pu)	
		Case 1 ^b	Case 2 ^c
.8 h	1.	4.2	2.6
.1 day	3.	4.4	2.9
.1 wk	21.	9.7	5.3
.2 wk	42.	17.8	7.1
.1 month	84.	34.8	9.7

^aDetection probability = 0.5, false-alarm probability = 0.001.

^bNo recalibrations within the accounting period, and 10% estimates of column inventories.

^cTwo-day recalibrations of input/output concentration and flow measuring instruments, and 5% estimates of column inventories.

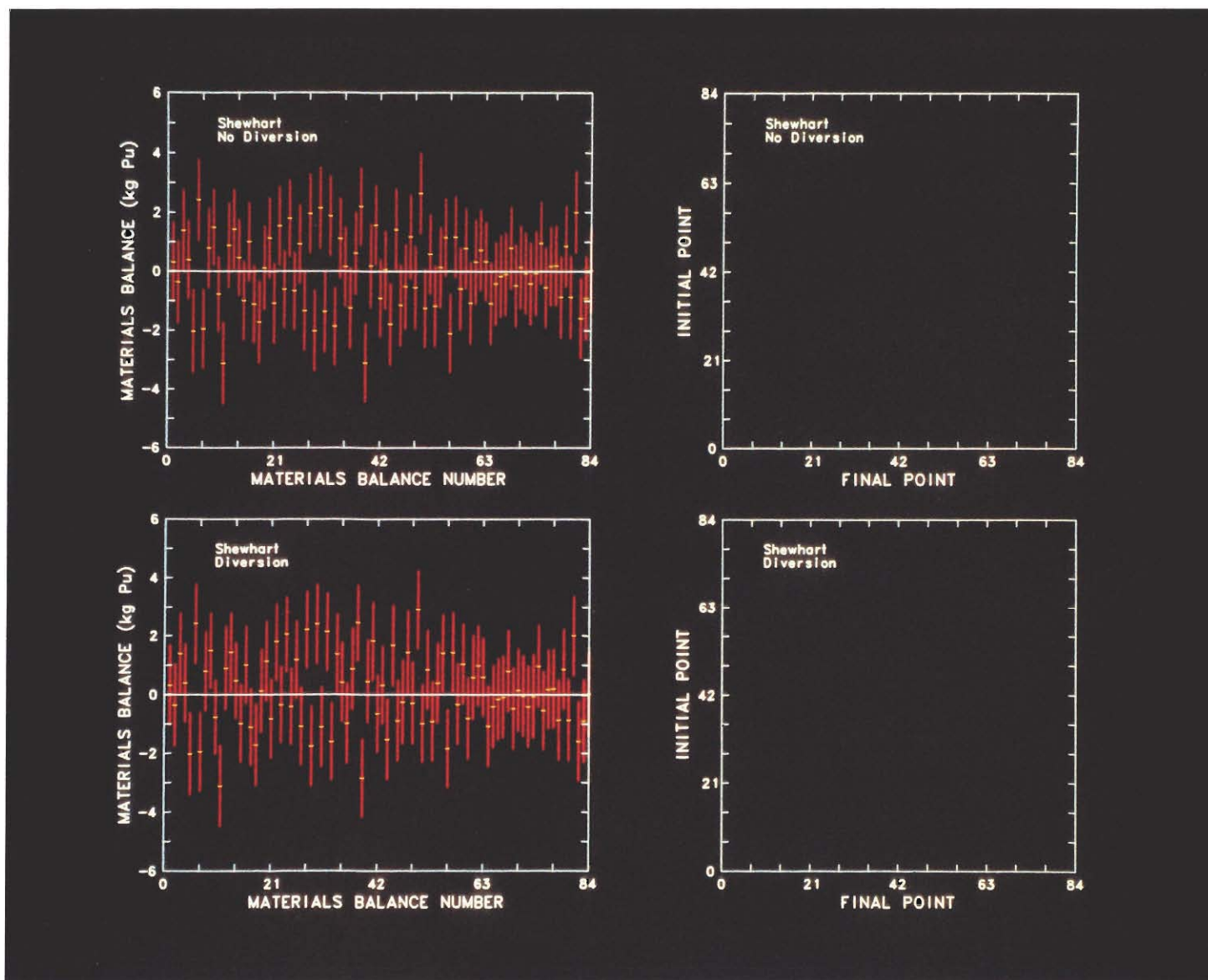


Fig. 14. No-diversion and diversion Shewhart charts with associated alarm charts. The diversion strategy is a low-level, uniform diversion of material between balances 21-63. That diversion is occurring is not obvious from the diversion Shewhart chart and no alarms show on the alarm chart.

tions, only one set of data will be available for making decisions, rather than the multiple data streams generated from a simulation. In particular, direct comparison of charts with and without diversion, as shown here, will be impossible. The decision-maker will have to extrapolate from historical information and from careful process and measurement analysis to determine whether diversion has occurred.

The results of the evaluation for two measurement strategies are given in Table VI. The diversion detection sensitivity for 1 week and less is limited by the uncertainties in the in-process inventory, which is both large (≈ 40 kg of plutonium) and difficult to measure. For

longer times, the sensitivity is limited by the systematic errors in the transfer measurements.

The short-term sensitivity to diversion could be improved by modifying equipment at the codecontamination-partitioning step. In the Purex process, plutonium and uranium are coextracted from the dissolver solution and then selectively extracted in what are called solvent-extractor contactors. In the reference facility, a series of pulsed-column contactors are used for the uranium-plutonium partitioning. These contactors have a relatively large plutonium inventory (≈ 25 kg), which not only varies under normal operating conditions but also is not amenable to ac-

curate measurement. Replacing the pulsed-column contactors with centrifugal contactors would decrease the plutonium inventory by an order of magnitude and improve the short-term (inventory-dominated) diversion sensitivity.

However, at about 1 month, the diversion sensitivity becomes throughput dominated; that is, errors in measuring the plutonium throughput determine the detection sensitivity. Even the best-case 1-month sensitivity (9.7 kg) seems rather large. However, the throughput of this facility is also large (1400 kg plutonium/month), so the sensitivity is 0.7% of throughput, which is really rather good. For this facility, though, im-

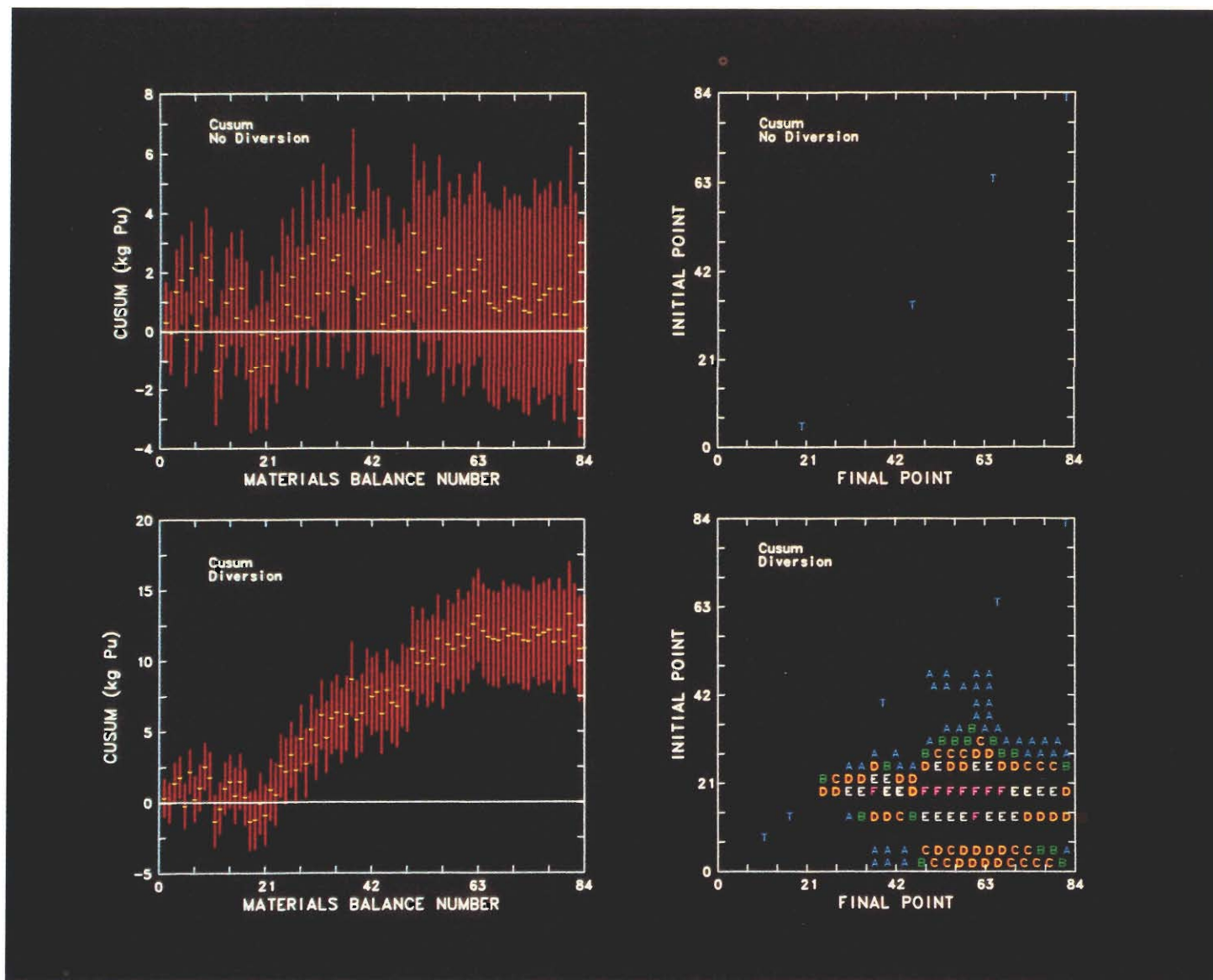


Fig. 15. No-diversion and diversion cusum charts with associated alarm charts. The diversion strategy is as described for Fig. 14. It is obvious from the diversion cusum chart that material is being diverted at about balance number 23. The cusum increases, indicating a continued diversion of material, until about balance number 63. Subsequently, the cusum maintains a roughly steady, high (≈ 12 -kg) value, indicating the total loss of a fixed quantity of material. To confirm these observations, the associated alarm charts begin to show alarms having small values of false-alarm probability ($\approx 10^{-3}$) at initial and final balance numbers of about 21 and 23. Balance numbers higher than 63 have high (≈ 0.5) false-alarm probability, which indicates that material is probably not being diverted.

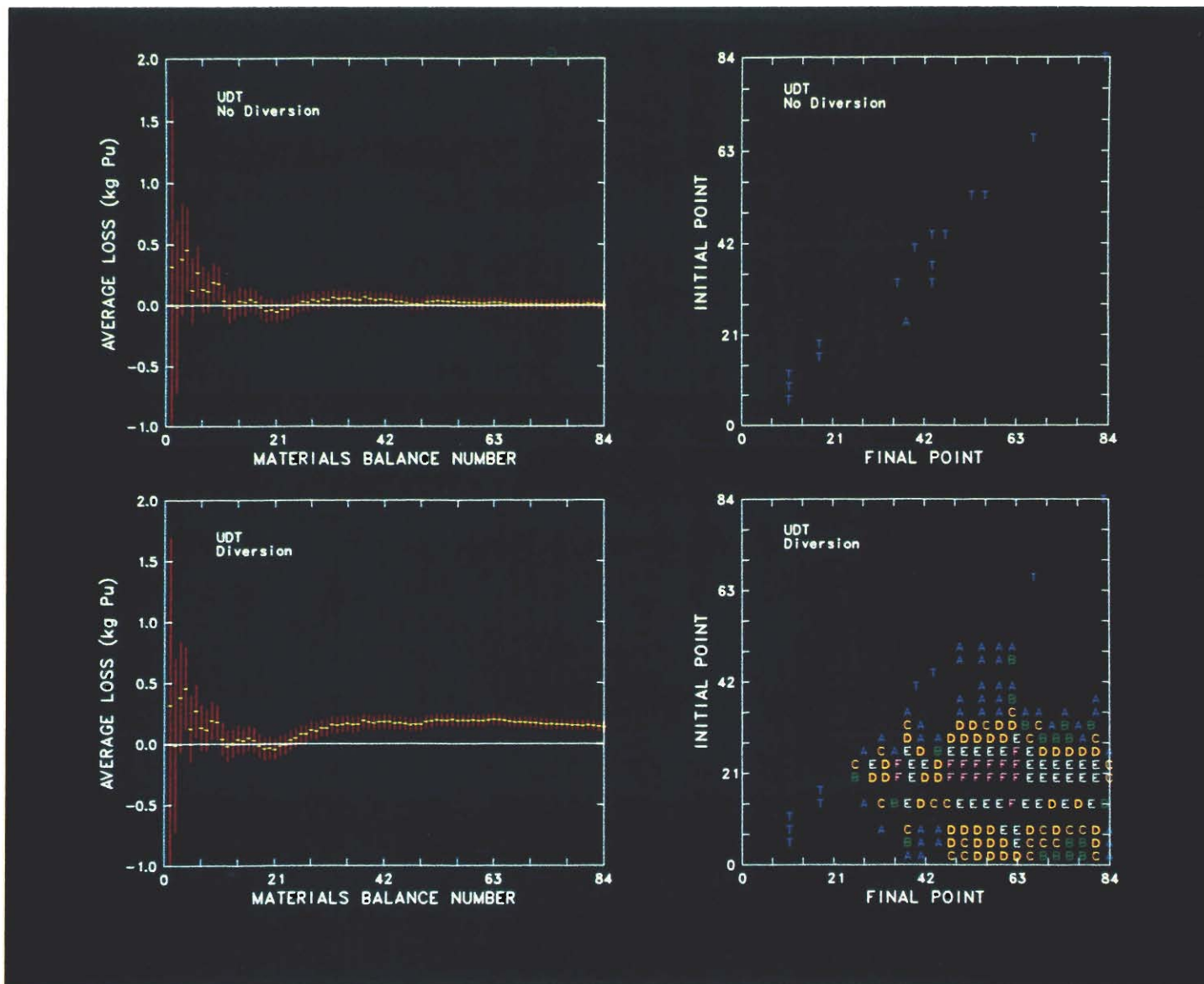


Fig. 16. No-diversion and diversion UDT charts with alarm charts. Again, the diversion strategy is as described for Fig. 14. The UDT diversion chart shows diversion commencing at about balance number 23, and the average material loss does not begin to decline until after number 63, when diversion has ceased. The alarm chart confirms these observations by the appearance of alarms at about balance numbers (21,23) and the absence of alarms in the vicinity of numbers (63,63).

provement in the long-term diversion sensitivity can be obtained only by better measurements of the throughput and better control of the correlated errors (such as calibration errors) in the throughput measurements.

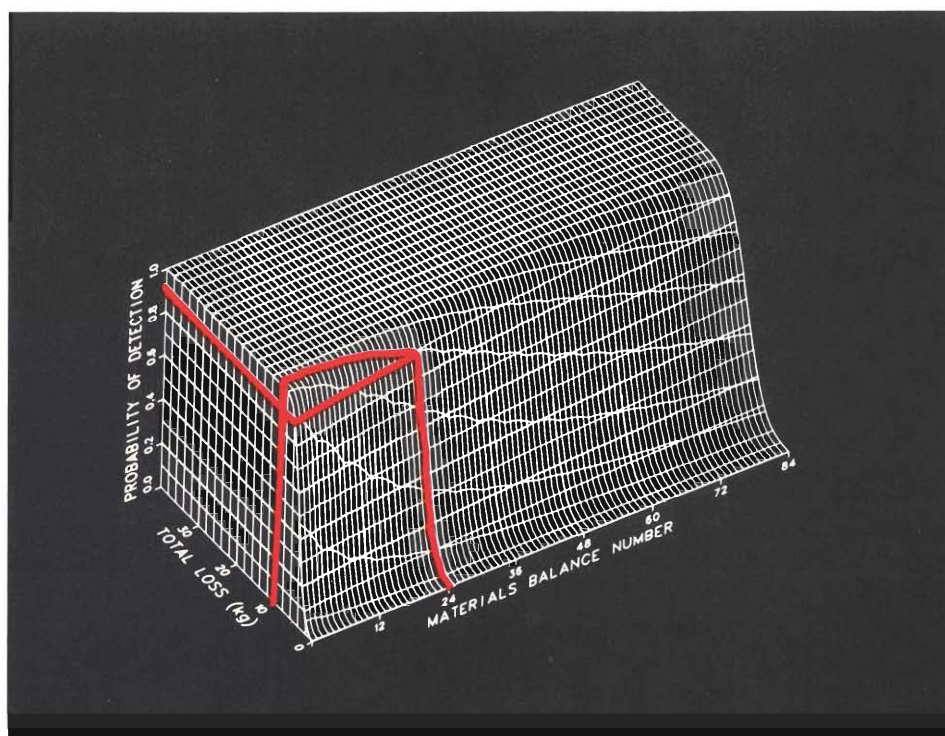
Figures 17a and 17b show examples of cusum performance surfaces from the simulated materials accounting data used to generate Table VI. Results for Case 1 (the worst case) are shown in Fig. 17a, and results for Case 2 (the best case) are shown in Fig. 17b. The figures illustrate the use of cusum performance surfaces in the design and evaluation of materials accounting systems. The improvement in sensitivity obtained by periodically recalibrating feed and product measuring devices is obvious when the figures are compared.

Discussion

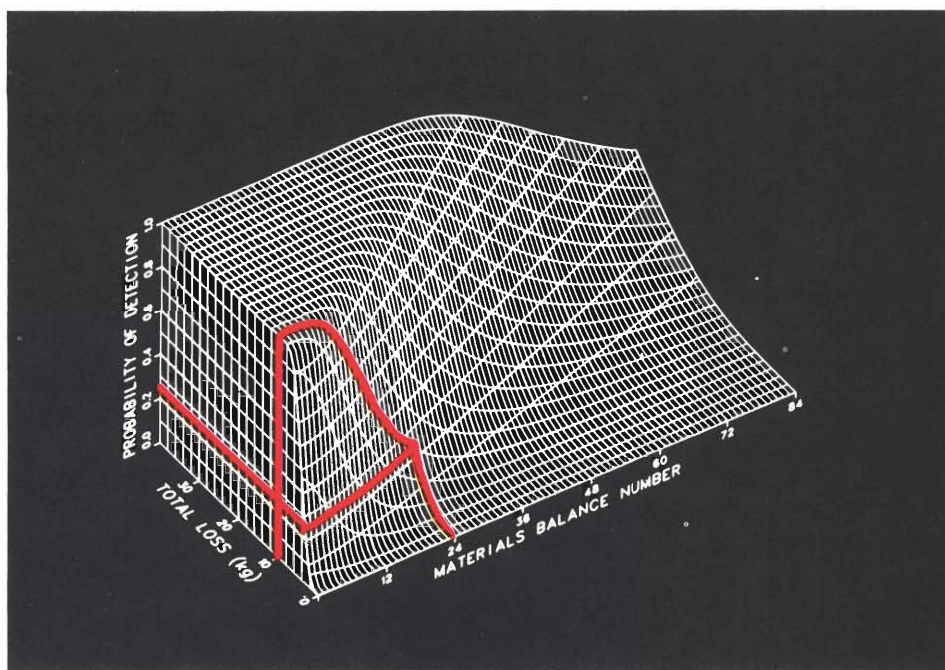
Until recently, almost no consideration was given to nuclear safeguards accounting requirements during the design of fuel-cycle facilities, the AGNS plant included. Instead the safeguards system designers were presented with either an existing facility or a relatively complete and fixed plant design. While the results of systems studies might introduce additional measurement instruments or bring about minor changes in operating equipment, they usually did not have any input to the choice of the process to be used in the facility or its mode of operation.

Increased recognition of the importance of nuclear safeguards and the need to integrate materials accounting into the process is bringing about a change. Safeguards designers are being consulted early in the design stages of fuel-cycle facilities. The resulting close cooperation between safeguards experts and process and facility designers should identify design alternatives that are both beneficial to safeguards and benevolent to the process.

The kind of materials accounting systems discussed above can provide better information on the locations and amounts of nuclear material than is



a.



b.

Fig. 17. Cusum performance surfaces for two accounting cases. In the worst case (b), the loss of 10 kg of material can be detected at the 24th materials balance with a probability of 0.25. For the best case (a), the loss of the same quantity of material at the 24th materials balance can be detected with a probability of 0.90. The importance of good measuring instruments and a good measurement program is clear.

currently provided by conventional methods. Such systems are beginning to be implemented at several facilities in the United States, including the new Los Alamos Scientific Laboratory Plutonium Facility, but much development work remains to be done. The process of com-

binning measuring instruments, data handling and analysis, and performance evaluation methodology into coherent effective safeguards systems is still in its infancy. The extension of these systems to international safeguards is just beginning.

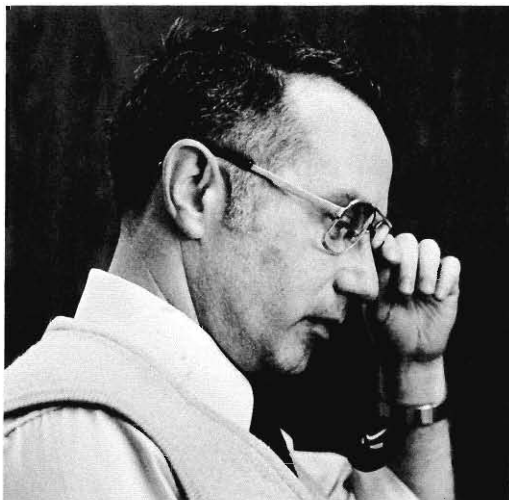
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Acknowledgment

The authors are indebted to their colleagues in Los Alamos Scientific Laboratory Group Q-4 and especially to H. A. Dayem, D. D. Cobb, and E. A. Hakkila for helpful discussions and for material from which portions of this article were adapted.

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